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FINAL REPORT

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Material Engineering of Novel Semiconductor Structures

Submitted to

the U.S. Air Force Office of Scientific Research
Program Manager - Dr. A. Craig

by

Principal Investigator:
Dr. J. Khurgin (Assistant Professor)

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RESULTS ACHIEVED SINCE INITIATION OF THE PROJECT (JUN 1 1989 - NOV 30 1990)

1. Experimental Investigation

Beginning in Summer'89 the nonlinear spectroscopy lab became operational. Two lasers were installed: visible Nd-YAG pumped dye laser and near-infrared Argon pumped dye laser. Also installed were liquid helium cryostat, 1000W Hg lamp, boxcar averager and lock-in amplifier.

The infrared laser had become fully operational near the end of 1989, therefore, our first experiments in the Fall of 1989 were done in the visible region of the spectrum, using familiar nonlinear material ZnSe in order to acquire experience in nonlinear spectroscopy [8] (three out of four graduate students working in the lab are new to the field). Highly sensitive measurements of the spectrum of nonlinear refractive index of ZnSe were done using the method of self-bending [15,19]. This method, developed by us, has shown significant advantage over conventional Kramers-Kronig method also performed by us [22].

The results of the measurements were somewhat unexpected, since the nonlinear index of refraction, n_2 measured by us turned out to be quite large ($10^{-8}\ cm^2/W$) and hard to explain using conventional wisdom of the thermally-induced bandgap shrinking. Therefore, we conducted additional set of measurements that gave the proof of the excitonic origin of the nonlinearity. These results had been recently reported in the Optics Letters[8].

After the near-infrared laser became fully operational, the main thrust of the experimental work was in the direction of investigation of nonlinear optical and electro-optical properties of the asymmetric coupled QW's. Efforts to locate reliable sources of MBE-grown QW material had finally succeeded. We have had samples *custom−grown* for us at University of California at Santa-Barbara (Prof. J. Merz group) and Martin-Marietta research labs. This collaboration is expected to continue. The characterization of the materials was done at the Applied Physics Laboratories, using Auger spectroscopy, Secondary Ion Mass Spectroscopy and scanning electron microscope.

Since the samples used in our research consist of $GaAs/Al_xGa_xAs$ epitaxial material grown on GaAs substrate, the substrate has smaller bandgap than the epitaxial layer. In order to study the absorption of the QW material, one needs to either etch away the substrate and perform the absorption measurements, or,

alternatively, perform photocurrent measurements. We have chosen the second option, since it allows us to bias the material and thus obtain a working electro-optical device. To prepare the samples for the photocurrent measurements, extensive processing was needed - etching, ohmic contacts evaporation, and wire bonding. The processing of the first set of samples had been done at Naval Research Labs and Harry Diamonds Lab. New samples are being processed at the Applied Physics Laboratory. We are planning, however, to have our own facilities by the end of 1990, including photoresist spinner, mask aligner, and two evaporators.

First experiments on photoluminescence and photoconductivity spectroscopy of asymmetric coupled QW's started in January 1990. The samples used by us had 25 coupled QW periods each of them consisting of two $Al_{0.35}Ga_{0.65}As$ QW's , with thicknesses of 18$\AA$ and 32$\AA$ separated by 15$\AA$ GaAs barrier. Thicker 100 $\AA$ barriers separated the coupled QW's from each other. The entire undoped multiple QW structure was sandwiched between n and p $Al_{0.35}Ga_{0.65}As$ layers, as shown in Fig.1. The whole structure thus represented a *pin* photodiode, with intrinsic layer being a multiple QW structure. In order to prevent the leakage, arrays of small photodiodes, 200μm in diameter were fabricated on mesa structures and ohmic contacts were evaporated.

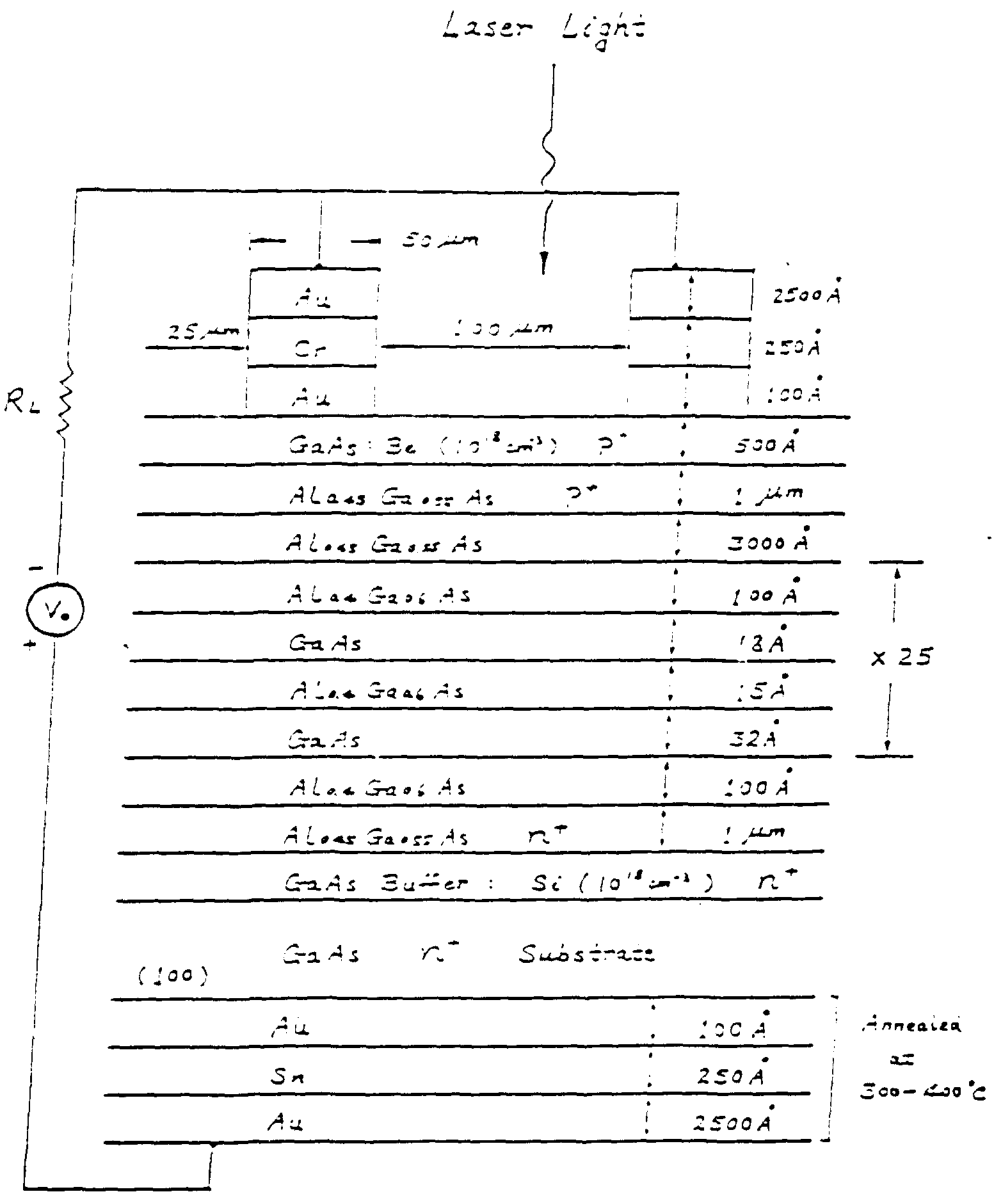

Fig. 1 Structure of p-i-n mesa diode in which intrinsic region contains asymmetric coupled quantum wells. See the text for detailed description of this structure. Photo-currents can be measured using this circuit with laser light shining on the sample.

The difference between our structure and all the structures reported by other researchers was in the way we place thicker QW in each period closer to the *n* side of the structure. As a result, when the reverse bias was applied, the *hole* energy levels in two coupled QW's could be brought into resonance, while most of the prior research[39,40] had concentrated on electron resonances.

Our preliminary calculations had shown that field of $\simeq 50 kV/cm$ was required to bring the hole energy levels of two wells into resonance and thus to increases the oscillator strength of the lowest excitonic transition. Since a built-in field of $\simeq 21 kV/cm$ was measured from I-V curve, the externally applied bias required for resonance was calculated to be about 2.1 V.

In Fig.2 the typical photocurrent spectra are shown. The sharp increase in peak photo current can be observed at the reverse bias equal to $2.25\,V$, indicating the resonance between the lowest hole levels in the ACQWs and thus confirming our earlier predictions [41]. Also, an extraordinary large blue shift of the resonance can be seen in Fig.2 The most unusual feature of the result is that the bias ranges at which the resonance occurred turned out to be much narrower than predicted. We are currently working on theoretical explanation of this feature (See Chapter 4.) One should also note the intensity dependence of the shifts, indicating the existence of the internal feedback loop in the circuit (Fig.3).

In Fig.4 the voltage dependence of the photocurrent is shown with clear indication of negative resistance region. Clearly, this kind of I-V behavior can result in optical bistability with the assistance of an external resistor (see Fig.1). Since the photocurrent depends on three parameters: laser intensity, laser wavelength, and bias, changing either one of these parameters, while keeping two other fixed should lead to optical switching and bistability. Indeed, we have achieved optical bistability in three distinct cases. [27–30]

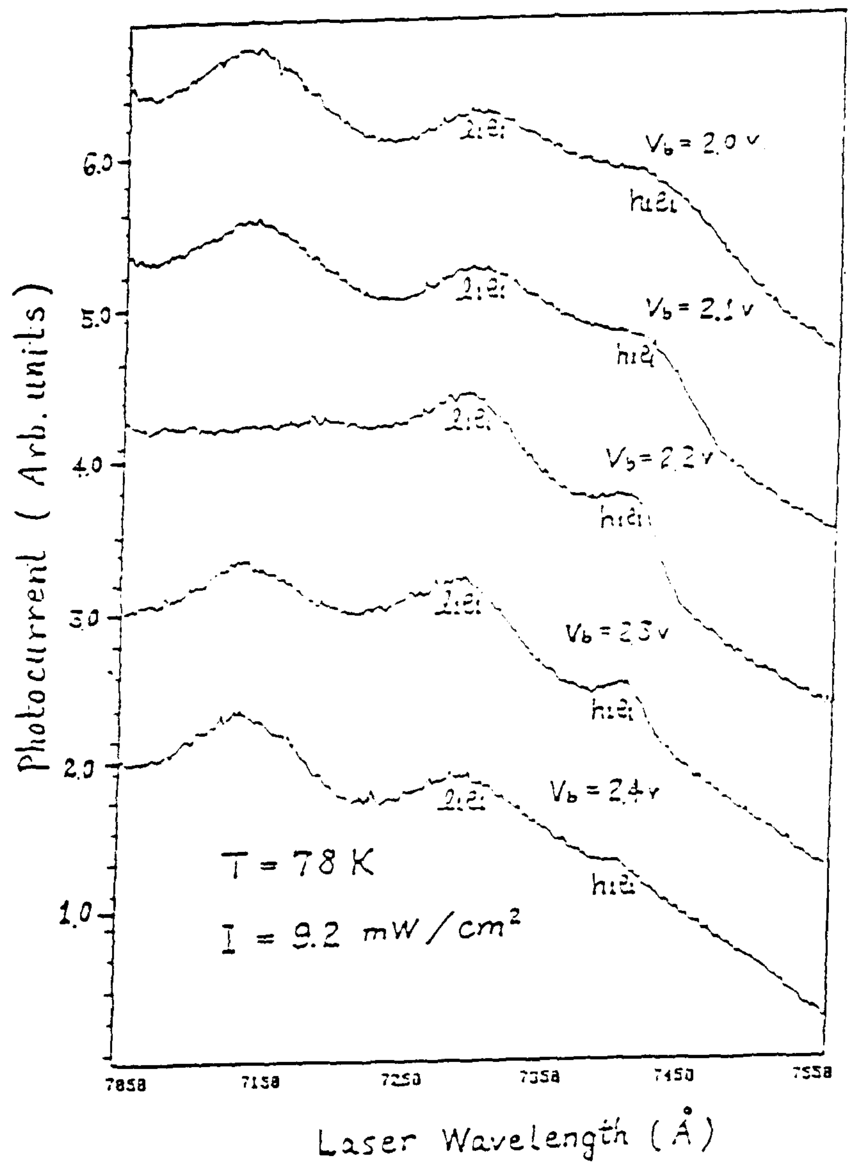

Fig. 2 The typical photo-current spectra are shown at the different (constant) reverse biases.

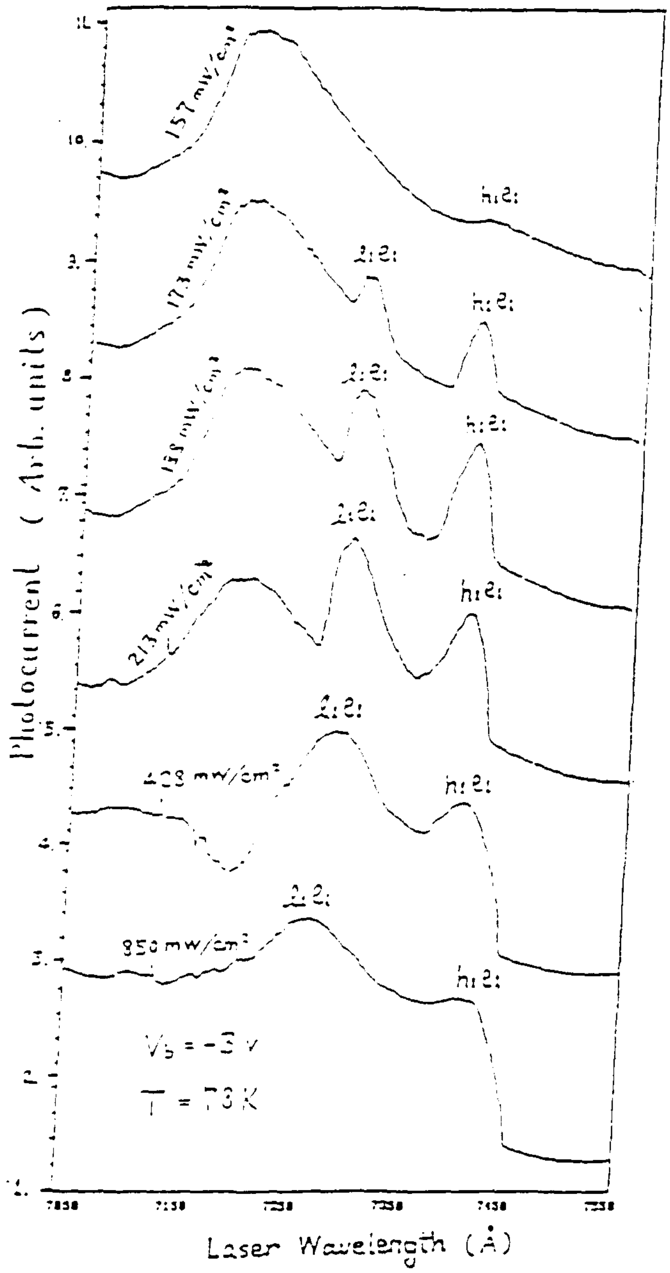

Fig. 3 The typical photo-current spectra are shown at the constant reverse bias $V_b \sim -3$ V for different laser intensities.

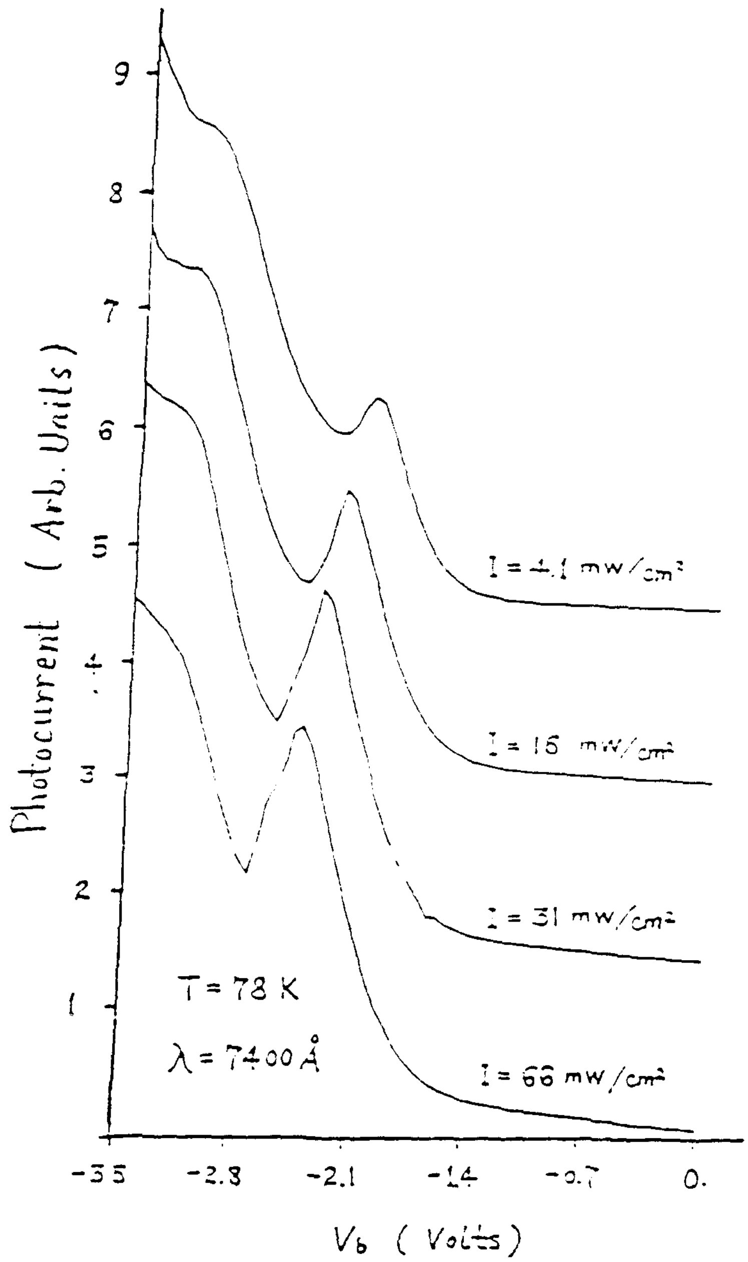

Fig. 4 Measurement of photocurrents vs. the reverse bias voltage for different laser intensities.

In Fig.5a typical photo-currents are measured vs. total bias in the circuit for a fixed laser intensity $I \simeq 58\ m\ W/cm^2$ and laser wavelength $\lambda \simeq 7411\ \AA$ with the external resistor $R_L \simeq 3.2\ M\,\Omega$. We can see from Fig.5a that optical switching (i.e. optical bistability and hysteresis) is demonstrated in this device.

In Fig.5b, typical photo-current is shown as a function of laser wavelength for the fixed bias $V_b \simeq -5.25\ V$ and laser intensity $I \simeq 64\ m\ W/cm^2$ with the external resistor $R_L \simeq 3.2\ M\,\Omega$. Huge hysteresis loop has been observed.

Finally In Fig.5c, photocurrent is shown as a function of laser intensity for a fixed laser wavelength $\lambda \simeq 7316\ \AA$ and total bias $V_b \simeq -5.5\ V$ with external resistor $R_L \simeq 3.2 M\,\Omega$. Once again, hysteresis is observed. It is worth noting that hysteresis loop in Fig.5c co-exists with the saturation of photocurrent indicating the large optical nonlinearity of the system.

The source of the blue shift larger than predicted is not yet known, and we are currently working on it. The results of our experiments had been presented at the Optical Society of America annual conference in November, 1990[27,28], at OSA topical conference on Quantum Optoelectronics [29] and will be published in the Optics Letters [10] in the Applied Physics Letters 11

In conclusion, the principal aim of the first stage of the experimental part of the project - observation of the strong nonlinear optical effects in asymmetric QW's have been achieved.

2. Study of excitonic effects in coupled QW's

The unexpected results of the experiments with coupled QW's have prompted us to initiate the in depth theoretical studies of coupled QW's. We have noticed, that in commonly used theoretical models the longitudinal (in the direction of growth) correlation of electron and hole wavefunctions is not taken into account. We have developed three new models that can adequately account for the longitudinal correlation. We have shown[9,21,23] that Coulomb attraction between electron and hole in coupled quantum wells leads to localization of states in the direction of growth, i.e. in one of the wells, even when the wells are symmetric and unbiased. As a result, oscillator strength of the lowest excitonic transition can increase by more than 100%, while the oscillator strengths of higher energy transitions can decrease. Besides shedding the light on the origin of the nonlinear effects in our experiments, these results, obtained using novel models, are important for understanding of the Wannier-Stark localization effect in quantum wells

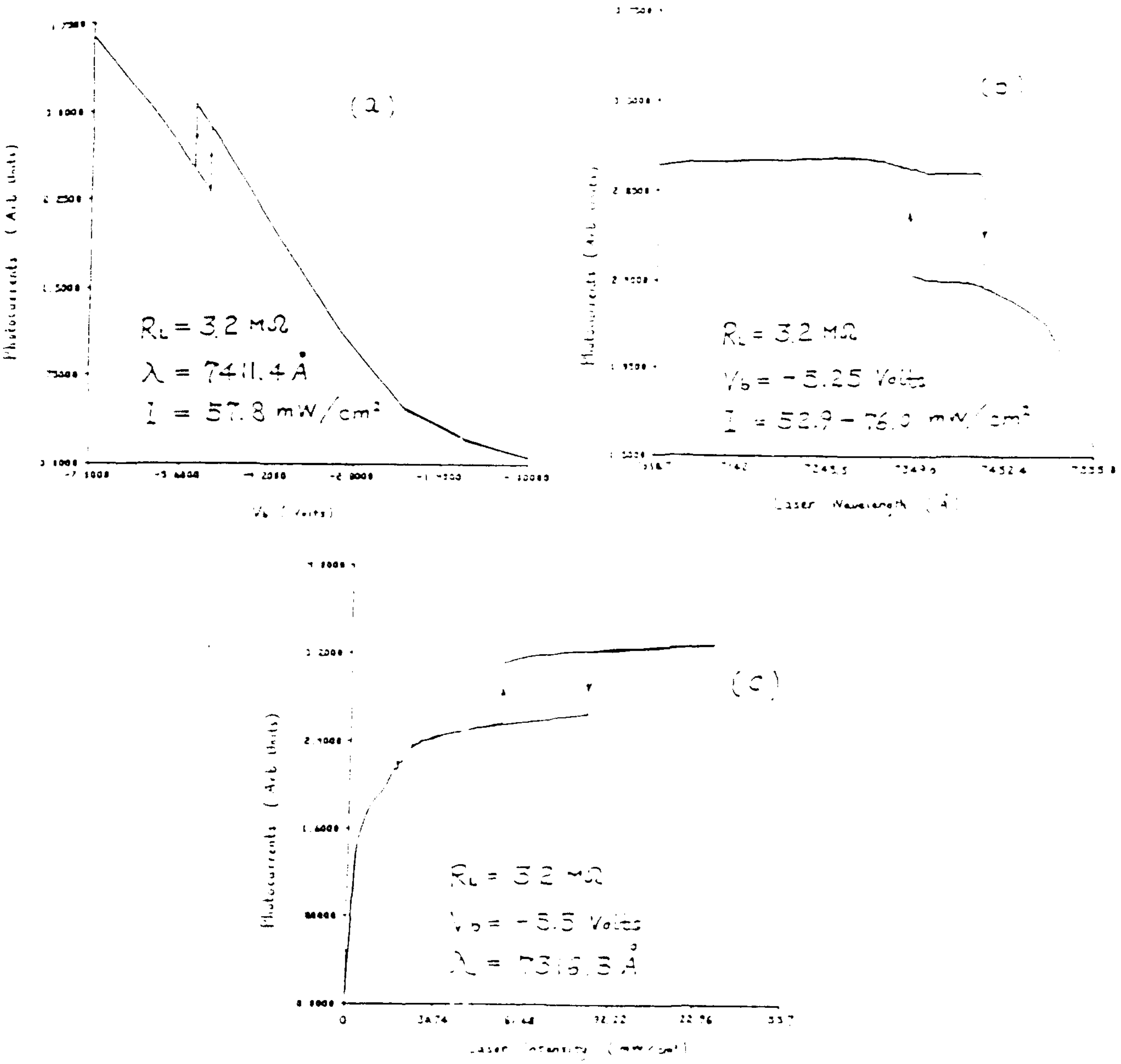

Fig. 5 With external feedback resistor $R_L \sim 3.2\,M\Omega$ typical photo-currents are measured vs. (*a*) total bias in the circuit for a fixed laser wavelength and intensity; (*b*) laser wavelength for a fixed bias and laser intensity; (*c*) laser intensity for a fixed bias and laser wavelength

and superlattices. The implications of the longitudinal coulomb correlations are discussed in depth in the upcoming publication in Physical Review B [10]. In the future we expect to study the exchange interaction in coupled QW's as well.

3. Study of intersubband second-order nonlinearities

A theory of second-order nonlinear susceptibility based on intersubband processes in asymmetric quantum well structures had been developed[3]. Large off-resonance nonlinearity had been predicted, and its origin had been explained.

We have also explained the reason for the huge difference (up to three orders of magnitude) between second order susceptibilities. For interband transitions, every state in the valence band is a ground state from which virtual transitions originate. The second order polarizations for the transitions originating from different ground states have different sums and effectively compensate each other. For the intersubband transitions only one ground state (lowest conduction subband) has carriers in it, therefore the compensation is not present.

The calculations have shown that second-harmonic coefficient in the $10\mu m$ range reaches $5\times10^{-9} m/V$ and Pockels coefficient $2\times10^{-8} m/V$. The dependence of second order nonlinear coefficients on bandgap offsets, effective masses, well geometries and doping concentration is investigated. The intersubband absorption was shown to be the main factor limiting the efficiency of quantum-well based nonlinear optical devices. The expressions for the absorption-limited efficiency of second-order nonlinear devices were obtained. The results show that asymmetric quantum well structures can be efficiently used in many infrared applications.

Recently, different groups of scientists have experimentally demonstrated the nonlinear effects predicted by us [37,38]. Both second harmonic generation and optical rectification have been observed confirming our theoretical results.

4. Proposals for novel bistable devices

Modulation-doped system of asymmetric coupled quantum wells was shown theoretically to possess a large optical nonlinearity due to the charge transfer associated with intersubband transition[2,18]. The range of parameters, leading to optical bistability has been determined. A novel triple quantum well system was

suggested, allowing a few orders of magnitude reduction in switching power density, to about $50\,W/cm^2$, while having response time of less than 100 ps. Optically-assisted transfer of up to 90% of all carriers between wells can become an operating principle of novel hybrid opto-electronic devices, incorporating the triple quantum well system into the channel of modulation-doped field-effect transistor with potential applications as fast opto-electronic switches and detectors in the 10 μm range.

A new scheme for achieving intrinsic optical bistability and switching in semiconductor heterostructures has been proposed [4,22]. The scheme does not require any external optical or electrical elements and possesses advantages of both the self electrooptic effect device and doping superlattices. This scheme is called "compensated" hetero-nipi superlattice and consists of asymmetric QW's placed in the intrinsic regions of hetero-nipi superlattice, and is called a hetero-nipi superlattice. Switching properties of the proposed device were theoretically analyzed and conditions for bistable operation were determined. First experimental structures have been designed and are being currently grown at UCSB.

5. Investigation of asymmetric quantum wells subjected to magnetic fields.

We have shown [6,17,20] that in the asymmetric quantum wells placed in the transverse magnetic field, optically excited carriers have asymmetric distribution in k - vector space, and therefore, they have finite drift velocity in the direction perpendicular to both growth direction and field direction. Depending on whether the external circuit is open or closed, it results in either voltage along the QW plane or the current in that direction. The voltage (current) pulse decays with the time constant equal to that of intraband relaxation time, i.e. of the order of $1ps$ or less. This may serve as a basis for future ultrafast detectors or as a tool for generation and study of hot carriers. Current sensitivity of the proposed device is of the order of few $\mu A/W$ at few Tesla magnetic fields.

6. Study of effects of stress on coupled quantum wells and wires.

New type of piezoelectric effect[1], based on strain-induced changes in confinement of the carriers in the asymmetric quantum well structures has been discovered and studied theoretically. This effect can exist in materials that in a bulk form possess a center of inversion symmetry and therefore do not exhibit conventional

piezoelectric properties. The magnitude of the piezoelectric coefficient in *GaAs/AlGaAs* structures was estimated to be few percent of that in quartz. The various schemes for its enhancement, including possible applications of quantum wires were proposed.

7. Research on coulomb enhancement of ultrafast nonlinearities.

Recently we have started looking for new ways to enhance the dynamical Stark effect by as much as an order of magnitude through the use of bandgap engineering. We have shown [25,28,29,30] that in QW's in which centers of mass of the electron and hole wavefunctions are separated by distances on the order of the exciton radius in the direction of growth, there exists a dynamic Coulomb potential which leads to pulsations in energy separation between levels and significant enhancement of the dynamic Stark effect. We propose to continue our work in this direction in order to develop optimal structures for ultrafast optical switching and also to consider influence of exchange and correlation interactions, and exciton-exciton interactions on the Coulomb - enhanced dynamic Stark effect. Our initial results will be published soon in the Journal of Optical Society of America B [12].

8. Study of frequency doubling in channel waveguides.

As mentioned above, successful progress had been made in that direction of calculating of second-order susceptibilities in asymmetric QW's. Theoretical calculations have been completed [3], and their results have been confirmed by the numerous experiments in other laboratories [37,38] It has been shown that extraordinary large second order susceptibilities exist in asymmetric QW structures. In order to effectively utilize large second harmonic coefficient of planar structures, we have initiated the investigation of the effective phasematching method. in planar or channel waveguides.

In the course of this work, a new method of phasematching in waveguides has been proposed by this principal investigator [7]. The method has been experimentally discovered at Philips Laboratories in ion-exchanged $KTiOPO_4$ (KTP) segmented waveguides made by Dupont. In this waveguides segmentation was achieved by using a mask during the process of ion exchange and thus fabricating short (few μm) segments of ion-exchanged waveguide separated by short segments

of nonguiding material. In this waveguides extraordinary high (more than 10%) frequency conversion efficiency of 880nm into 440 nm radiation has been achieved, using the largest (d_{33}) SHG coefficient of KTP. Subsequent investigation has pinpointed the cause of this extraordinary result: quasiphasematching *achieved by strong modulation of phase mismatch between fundamental and second harmonic radiation* It has been shown that the phase mismatch in guiding segment is much larger than in nonguiding segment, and, as a result, there is a grating of phase mismatch. This grating causes an increase in SHG efficiency comparable to the increase due to the reversal of ferroelectric domains. Our findings, reported in the upcoming issue of Applied Physics Letters , lead to the conclusion, that by simply growing alternative segments of the materials with large and small phase mismatch, high SHG efficiency can be achieved. We plan to use our results to achieve high frequency conversion efficiency in $ZnSe-ZnS_xSe_{1-x}$ waveguides grown by MBE and MOCVD.

9. Study of Optically Pumped Double Heterostructure visible lasers.

Availability of $ZnSeZnS_xSe_{1-x}$ double heterostructure waveguides with high degree of both light and carrier confinement has prompted us to start the optical pumping of these samples. Using third harmonic of NdYAG laser as a pump radiation, stimulated emission had been achieved in a dozen of samples with various compositions and geometries, including superlattices, single QW's and so on. The results are to be reported in the upcoming publications [13,31]. We are currently working on optimization of the heterostructure design for the lowest lasing threshold and the highest slope efficiency.

PUBLICATIONS BY OUR GROUP SINCE THE INITIATION OF THE GRANT (June,1989 - Feb,1991)

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1. J. Khurgin, "Quantum-confined piezoelectric effect" *J. Appl. Phys.*, **66**, 994, (1989)

2. J. Khurgin "Electro-optic Switching and Bistability in Coupled Quantum Wells", *Appl. Phys. Lett.*, **54**, 2589, (1989)

3. J. Khurgin, "Second Order Intersubband Nonlinear Optical Susceptibilities of Asymmetric Quantum Well Structures", *J. Opt. Soc. Am.* **B**, **6**, 1673, (1989)

4. J. Khurgin "Intrinsic optical bistability in compensated hetero-nipi superlattices" *IEEE J. Quantum Electron.*, **QE-26**, 876, (1990)

5. S. Colak and J. Khurgin, "Stability of Frequency Doubling Phase Matched with II-VI Microcrystals", *Journal of Crystal Growth* **101**, 748, (1990)

6. J. Khurgin "Optical Generation of Picosecond Electrical Pulses in Asymmetric Quantum Well Structures Placed in Transverse Magnetic Field". *Appl. Phys. Lett.*,, **56**, 2490 (1990)

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Pending:

9. S. Li, and J. Khurgin, "Longitudinal Coulomb Attraction in Coupled Quantum

Wells" submitted to *Phys. Rev.* **B.** in Dec. 1990

10. C. L. Guo, Y. J. Ding, S. Li, J. B. Khurgin, K.-K. Law, J. Stellato, C. T. Law, A. E. Kaplan, and L. A. Coldren, "Strong excitonic nonlinearity in a *pin* diode incorporating narrow asymmetric coupled quantum wells." submitted to *Opt. Lett.*

11. Y. J. Ding, C. L. Guo, S. Li, J. B. Khurgin, K.-K. Law, J. Stellato, C. T. Law, A. E. Kaplan, and L. A. Coldren, "Observation of anomalously large blue shift of the excitonic transition and optical bistability in narrow as,mmetric coupled quantum wells," submitted to *Appl. Phys. Lett.*,

12. J. Khurgin, "Coulomb enhancement of ultrafast nonlinearities in quantum wells", submitted to *J. Opt. Soc. Am.* **B**, Mar 1990

13. G. Sun, K. Shazad, J. Khurgin, J. Petruzello, and J. Gaines, "Light and Carrier confinement in the optically pumped double-heterostructure $ZnSe/ZnS_xSe_{1-x}$ lasers, submitted to *Appl. Phys. Lett.*,, Feb 1990

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14* S. Colak and J. Khurgin, "Stability of Frequency Doubling Phase Matched with II-VI Microcrystals", presented at Fourth International Conference on II-VI Compounds, Berlin Sept 17-22 1989

15. Y. J. Ding, G. A. Swartzlander Jr., J. B. Khurgin, and A. E. Kaplan, "Non-linear beam propagation effects of a pulsed laser beam in ZnSe." presented at 1989 Annual Meeting of Optical Society of America.

16 J. Khurgin, "Electro-optic switching in modulation-doped Coupled Quantum Wells", presented at 1989 Annual Meeting of Optical Society of America.

17 J. Khurgin, "Optical generation of picosecond electrical pulses in asymmetric quantum well structures placed in transverse magnetic field", presented at 1989 Annual Meeting of Optical Society of America.

18* J. Khurgin, "Nonlinear properties of asymmetric quantum well structures", to presented at International Conference on Lasers (Lasers'89), December 3-8, 1989 (invited talk)

19* Y. J. Ding, C. L. Guo, G. A. Swartzlander, J. B. Khurgin, and A. E. Kaplan, "Multimethod measurement of nonlinear refractive index in ZnSe", presented at International Conference on Lasers (Lasers'89), December 3-8, 1989

20* J. Khurgin "Ultrafast Laser Probing of Asymmetric Quantum Well Structures in Transverse Magnetic Field.", presented at SPIE symposium on Ultrafast Laser Probe Phenomena in Bulk and Microstructure Semiconductors, 18-19 March, 1990, San Diego, Ca, and published in the "*Proceedings of SPIE*", **1282,** p. 203 (1990)

21* J. Khurgin, S. Li "Longitudinal Coulomb Attraction in Coupled Quantum

* - Refereed presentation

Wells" presented at SPIE symposium on *Quantum Well and Superlattice Physics*, San Diego 18-19 March, 1990 and published in the "*Proceedings of SPIE*", **1283**, p. 317 (1990)

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23* S. Li, J. Khurgin, " Enhancement of the Oscillator Strength in Coupled Quantum Wells due to Longitudinal Coulomb Attraction" CLEO'90 Technical Digest, Anaheim, Ca May 1990 p.84

24*. J. Khurgin "Intrinsic optical bistability in asymmetric hetero-nipi superlattices" CLEO'90 Technical Digest, Anaheim, Ca May 1990 p.144

25* J. Khurgin "Coulomb enhancement of dynamic Stark effect in quantum well structures". IQEC'90 postdeadline papers, Anaheim, Ca, May 1990 p. 379

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29* Y. J. Ding, C. L. Guo, S. Li, J. B. Khurgin, K-K Law, J. Stellato, C. T. Law, A. E. Kaplan, and L. A. Coldren, "Large excitonic blue-shift and nonlinearity in narrow asymmetric coupled quantum wells", *presented at* OSA topical meeting on Quantum Optoelectronics", Salt Lake City, Mar 11-13, 1991 and published in the

proceedings, p. 79

30* J. Khurgin, "Coulomb enhancement of ultrafast nonlinearities in semiconductor quantum wells", presented at OSA topical meeting on Quantum Optoelectronics", Salt Lake City, Mar 11-13, 1991 and published in the proceedings, p. 102

Pending:

31* G. Sun, K. Shazad, J. Khurgin, and J. Gaines, "Low threshold optically pumped double-heterostructure $ZnSe/ZnS_xSe_{1-x}$ lasers", accepted to be presented at CLEO-91,

32* Y. J. Ding, C. L. Guo, S. Li, J. B. Khurgin, K-K Law, J. Stellato, C. T. Law, A. E. Kaplan, and L. A. Coldren, "Observation of anomalously large blue shift and optical bistability in narrow asymmetric coupled quantum wells", accepted to be presented at CLEO-91

33* C. L. Guo, Y. J. Ding, S. Li, J. B. Khurgin, K-K Law, J. Stellato, C. T. Law, A. E. Kaplan, and L. A. Coldren, "Evidence of the intrinsic feedback in photocurrent response of *pin* diode incorporating narrow asymmetric coupled quantum wells", accepted to be presented at CLEO-91

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